Resistivity studies under hydrostatic pressure on a low-resistance variant of the quasi-2D organic superconductor κ-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Br: quest for intrinsic scattering contributions

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Resistivity measurements have been performed on a low (LR)- and high (HR)-resistance variant of the κ-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Br superconductor. While the HR sample was synthesized following the standard procedure, the LR crystal is a result of a somewhat modified synthesis route. According to their residual resistivities and residual resistivity ratios, the LR crystal is of distinctly superior quality. He-gas pressure was used to study the effect of hydrostatic pressure on the different transport regimes for both variants. The main results of these comparative investigations are (i) a significant part of the inelastic-scattering contribution, which causes the anomalous $\rho(T)$ maximum in standard HR crystals around 90 K, is sample dependent, i.e. extrinsic in nature, (ii) the abrupt change in $\rho(T)$ at T* $\approx$ 40 K from a strongly temperature-dependent behavior at T > T* to an only weakly T-dependent $\rho(T)$ at T < T* is unaffected by this scattering contribution and thus marks an independent property, most likely a second-order phase transition, (iii) both variants reveal a $\rho(T) \propto AT^2$ dependence at low temperatures, i.e. for T$_c$ $\leq$ T $\leq$ T$_0$, although with strongly sample-dependent coefficients A and upper bounds for the T^2 behavior measured by T$_0$. The latter result is inconsistent with the T^2 dependence originating from coherent Fermi-liquid excitations.

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I. INTRODUCTION

Organic charge-transfer-salts, based on the electron-donor molecule BEDT-TTF (bisethylenedithiotetrathiafulvalene) — or simply ET — form layered structures consisting of alternating sheets of conducting (ET)$_2^+$ cations and insulating anions X$^-$. Within this class of materials, the κ-phase (ET)$_2$X salts with X = Cu[N(CN)$_2$]Cl, Cu[N(CN)$_2$]Br and Cu(NCS)$_2$ are of particular interest due to the variety of electronic phases encountered as a function of hydrostatic pressure or anion substitution. According to the conceptual phase diagram proposed by Kanoda, the ground state of the system is controlled by the parameter W/U$_{eff}$, i.e., the width of the conduction band W relative to the effective on-site Coulomb repulsion U$_{eff}$, a ratio which can be changed by hydrostatic pressure or chemical substitutions[1]. This conceptual phase diagram implies that the antiferromagnetic insulator X = Cu[N(CN)$_2$]Cl and the correlated metal X = Cu[N(CN)$_2$]Br lie on opposite sites of a bandwidth-controlled Mott-transition. The region across this metal-to-insulator transition has been explored in great detail by employing pressure studies of various magnetic[2], transport[3,4] and acoustic[5] properties. These studies confirm earlier results[6,7] which revealed that at a pressure of 300-400 bar, i.e., above the critical region of coexistence of insulating and metallic phases[2,4], the X = Cu[N(CN)$_2$]Cl salt shows the same highly unusual resistivity profile $\rho(T)$ as the Cu[N(CN)$_2$]Br system at ambient pressure[8]. Three distinct transport regimes have been identified[4]: (i) a *semiconducting* high-temperature range, (ii) a *bad metal* behavior at intermediate temperatures with a strongly temperature-dependent $\rho(T)$ and a pronounced maximum around 90 K that marks the crossover to regime (i), and (iii) a $\rho \propto AT^2$ behavior at low temperatures preceding the superconducting transition at T$_c$. Various explanations have been proposed for the different transport regimes. Suggestions for the anomalous resistance maximum include an order-disorder-transition of the ethylene endgroups of the ET molecules[9,10,11] and a crossover from localized small-polaron to coherent large polaron behavior[12], see also[13]

for earlier arguments on the resistance anomaly. Alternatively, the "bad metal" regime (ii) together with the T^2 dependence at low temperatures have been linked to the strongly correlated nature of the electrons[4,14]. Within a dynamical mean-field (DMFT) approach, Merino et al.[14] predicted a smooth crossover from coherent Fermi-liquid excitations with $\rho \propto T^2$ at low temperatures to incoherent (*bad metal*) excitations at higher temperatures. Using such DMFT calculations for a simple Hubbard model, Limelette et al.[4] recently attempted to provide even a quantitative account for the $\rho(T)$ behavior of pressurized X = Cu[N(CN)$_2$]Cl over an extended temperature range covering almost all three of the above-cited transport regimes (i)-(iii).

It is fair to say, however, that despite the intensive efforts from both experimental and theoretical sides to explain the anomalous state above T_c, its nature still remains puzzling. In that respect, a deeper understanding of the unusual $\rho(T)$ behavior would be of paramount importance given that the inelastic-scattering mechanism, which causes the electrical resistivity of a superconductor above T_c, is usually identical to the relevant pairing interaction.

In this paper, we report resistivity measurements on a low (LR)- and standard high(HR)-resistance variant of the X = Cu[N(CN)$_2$]Br superconductor. These comparative studies, which include measurements under hydrostatic pressure, disclose striking differences between both variants. Our results demonstrate that — in contrast to conventional metals obeying Matthiessen's rule — extrinsic factors such as disorder, defects or impurities may strongly affect the inelastic scattering contribution in the present molecular conductors.

II. EXPERIMENTAL

The temperature dependence of the resistivity was measured by employing a standard four-terminal ac-technique operating at a frequency of 17 Hz. A maximum current of 10 μA was used to avoid self-heating. The electrical contacts to the crystal were made by 25 μm Cu wires attached to the sample by graphite paste. Typical contact resistances were $\leq 10~\Omega$. Owing to the large in-plane vs. out-of-plane resistivity anisotropy in these materials and the irregular shape of the crystals, an accurate determination of the in-plane resistivity $\rho_\parallel$ is very difficult, see e.g.[15,16,17]. As pointed out in these references, those in-plane data derived from a standard measurement geometry with four contacts on the same face of the crystal, almost always contain a significant interlayer component $\rho_\perp$. Thus, most reliable resistivity data, free of such mixing effects, can be obtained from out-of-plane measurements. To rule out errors which might originate in an inhomogeneous current flow in our four-terminal out-of-plane measurements, comparative investigations using a six-terminal configuration were conducted and found to deviate by not more than 4 % at maximum. For

the latter measurement geometry, the current had been fed through the crystal by two pairs of terminals (the outer two of three terminals) attached to opposite crystal surfaces assuring these surfaces to be equi-potential planes. These $\rho_\perp$ data enable even a quantitative comparison with corresponding results on other crystals to be performed. For the in-plane measurements, $\rho_\parallel$, the current contacts were placed on opposite end surfaces of the crystal. A He-gas-pressure technique was used to ensure hydrostatic pressure conditions. The measurements were performed at a low sweep rate of 0.1 K/min to guarantee thermal equilibrium.

The HR single crystal of κ(BEDT-TTF)$_2$Cu[N(CN)$_2$]Br was synthesized at the Argonne National Laboratory following the standard procedure as described elsewhere[18]. The LR crystal was obtained by solving 60 mg BEDT-TTF, 80 mg tetraphenyl-phosphoniumdicyanamid (Ph$_4$PN(CN)$_2$) and 20 mg CuBr in a mixture of 80 ml tetrahydrofuran (THF) and 20 ml ethylenglycol (EG). The solution was filled in a three-chamber electrolyte cell. The crystals were then grown at a current of 35 μA and a voltage of 1.3 V applied over a period of 14 days.

III. RESULTS

Figure 1 shows resistivity profiles $\rho(T)$ for the LR and HR κ-(ET)$_2$Cu[N(CN)$_2$]Br single crystals at various pressures. The data have been normalized to their room-temperature values taken at ambient pressure. The figure discloses striking differences in the charge transport for both variants: Instead of the semiconducting increase in $\rho_\perp$ at higher temperatures and the pronounced maximum around 90 K that characterizes standard samples and is also present in the HR crystal studied here (Fig. 1c), the LR crystal (Fig. 1a) remains metallic below 300 K. Rather its resistivity profile reveals a weak reduction upon cooling down to about 120 K, below which it starts to drop more rapidly. This shoulder near 100 K is likely to be a remnant of the resistivity hump in the HR sample. In fact, as can be seen in Fig. 1b, a maximum around 100 K shows up in the in-plane resistivity data for the LR crystal as well. For the resistivity anisotropy, $\rho_\perp/\rho_\parallel$, our measurements reveal a lower limit of about 100 at room temperature.

Apart from these sample-dependent contributions, the $\rho(T)$ data for both crystals exhibit a sharp dip at $T_g = 77$ K. This anomaly has been assigned to a glass transition associated with a freezing of orientational degrees of freedom of the ethylene endgroups[19,20]. With increasing pressure, the out-of-plane resistivity for both crystals becomes substantially reduced. This effect is most strongly pronounced at intermediate temperatures 40 K $\lesssim$ T $\lesssim$ 200 K, with a relative reduction $\rho^{-1}\Delta\rho/\Delta p = \rho(T, p_0 = 0)^{-1}[\rho(T, p_0) - \rho(T, p)]/(p_0 - p)$ for p = 170 bar corresponding to about -(360 $\pm$ 20) %/kbar at 50 K and -(180 $\pm$ 15) %/kbar at 80 K for the HR crystal. A somewhat
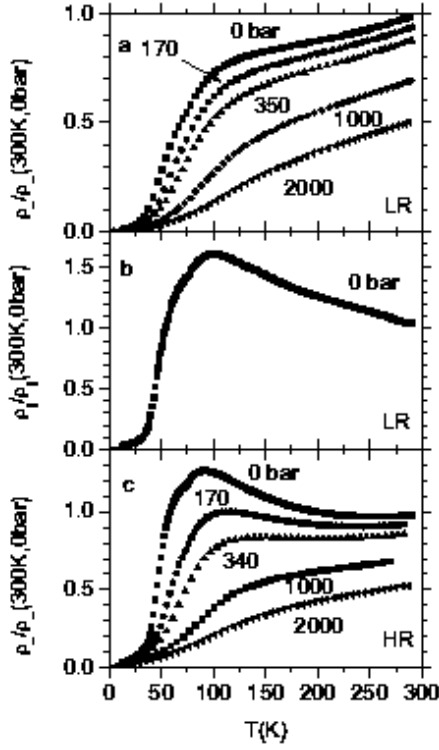
FIG. 1: Temperature dependence of the resistivity of single crystalline κ-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Br at various hydrostatic pressure values up to 2000 bar. Measurements were performed on the low-resistance (LR) crystal with current perpendicular (a) and parallel (b) to the highly conducting planes and for a standard high-resistance (HR) sample perpendicular to the planes (c).
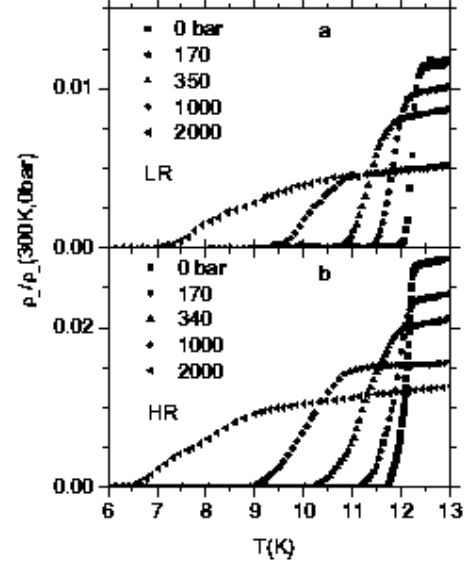

FIG. 2: Low temperature out-of-plane resistivity data at various hydrostatic pressure values up to 2000 bar for the low (LR)(a)- and high (HR)(b)- resistance variant of κ-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Br.

strongly temperature-dependent behavior at intermediate temperatures to an only weakly temperature dependent $\rho(T)$ at low temperatures.
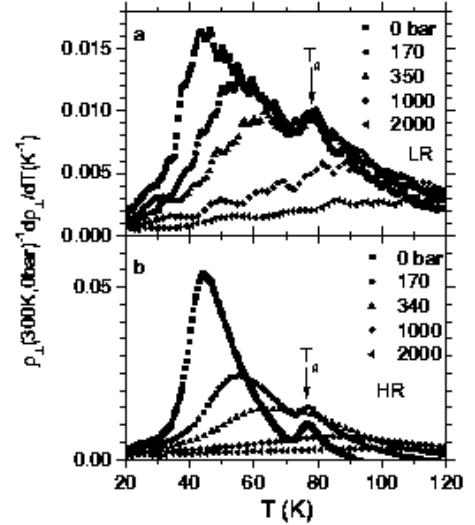

FIG. 3: Temperature derivative of the out-of-plane resistivity data for the low (LR) (a)- and high (HR)(b)- resistance variants of κ-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Br at various pressures. Arrows indicate the glass-transition temperature at T_g associated with frozen-in disorder of the ethylene endgroups.

smaller, though still very large, pressure response of -(250 $\pm$ 20) %/kbar (50 K) and -(120 $\pm$ 10) %/kbar (80 K) is found for the LR crystal. At higher temperatures, i.e., T = 200 K and 250 K, the effect of pressure becomes substantially reduced reaching values of -(45 $\pm$ 5)%/kbar and -(35 $\pm$ 5) %/kbar, respectively, which is about the same for both crystals.

Figure 2 shows the low-temperature out-of-plane resistivity data for both crystals on expanded scales. For the LR crystal (Fig. 2a), the midpoint (50 % point) of the resistivity drop at ambient pressure is at 12.2 K with a 10–90 % width of only 0.2 K. With increasing pressure, the transition shifts to lower temperatures and broadens progressively. Using the midpoint as a measure of T_c, we find an initial pressure coefficient of $dT_c/dp|_{p\to 0}$ = -(2.6 $\pm$ 0.2) K/kbar. These numbers have to be compared with T_c = 12.0 K, a 10–90 % width of 0.4 K and $dT_c/dp|_{p\to 0}$ = -(2.4 $\pm$ 0.2) K/kbar for the HR crystal (Fig. 2b). The pressure coefficient of T_c for both crystals is in excellent agreement with the results of previous pressure studies yielding pressure coefficients of -2.4 K/kbar[6] and -2.8 K/kbar[7].

Common to the data sets for the LR and HR crystals in Fig. 1 is the almost abrupt change in $\rho(T)$ from a

This becomes even more clear in Fig. 3 where the derivative $d\rho_\perp/dT$ is plotted for the LR (Fig. 3a) and HR (Fig. 3b) crystals at different pressure values. For both samples, we find a pronounced maximum in $d\rho/dT$ at about the same temperature T_{max} = 44 K in accor-

dance with previous results on an HR crystal[21]. With increasing pressure, the maximum becomes reduced in size, rounded and shifted to higher temperatures. At a pressure of p = 2 kbar, the maximum has been suppressed almost completely. The sharp peak at the high-temperature side of the $d\rho/dT$ maximum in Fig. 3 reflects the glass transition. Its position is almost identical for both crystals with $T_g = 77$ K. With increasing pressure, the signature of the glass-transition becomes weaker while its position remains almost unaffected up to p = 170 bar and, for the LR crystal, even up to p = 350 bar. The data yield an upper limit for the pressure coefficient of T_g of $dT_g/dp|_{p\to0} \geq$ -0.6 K/kbar. At higher pressures $p \geq 1$ kbar, however, no indication of the glass transition can be resolved any more.

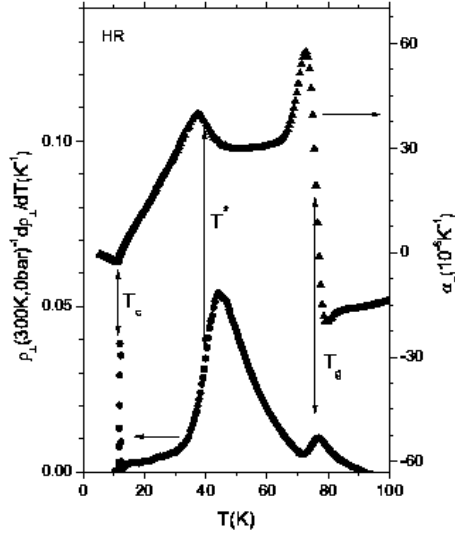FIG. 4: Temperature derivative of out-of-plane resistivity data, $d\rho_\perp/dT$, (left scale) and thermal expansion results (right scale) taken from reference [20] for two different high-resistance κ-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Br crystals plotted on the same temperature scale. The resistivity data for T $\leq$ 10.5 K have been omitted for clarity. Arrows indicate positions of the superconducting (T_c) and glass transition (T_g), as well as for the anomaly at T^*.

In Fig. 4 we compare the temperature dependence of the $d\rho_\perp/dT$ data of Fig. 3b with those of the coefficient of thermal expansion measured along the in-plane a-axis, α_a, on a similar HR crystal[20]. The figure discloses a clear correspondence of the features in $d\rho/dT$ with the phase-transition-like anomalies observed in $\alpha_a(T)$ at $T_c = 12$ K, $T_g = 77$ K and $T^* \simeq 40$ K[20]. More precisely, as indicated by the arrow at T^*, it is the midpoint of the low-T side of the $d\rho/dT$ maximum which coincides with the transition temperature T^* determined from $\alpha(T)$. Using the midpoint as a measure of T^*, the data in Fig. 3 can be used to determine the pressure dependence of T^*. For pressures p $\leq$ 350 bar, this criterion yields about the same pressure coefficient of $dT^*/dp|_{p\to0} = + (35 \pm 7)$ K/kbar for both variants. This value slightly exceeds the pressure

effect of about + 25 K/kbar reported by Frikach et al.[22] who followed the position of the pronounced minimum in the sound velocity as a function of pressure.
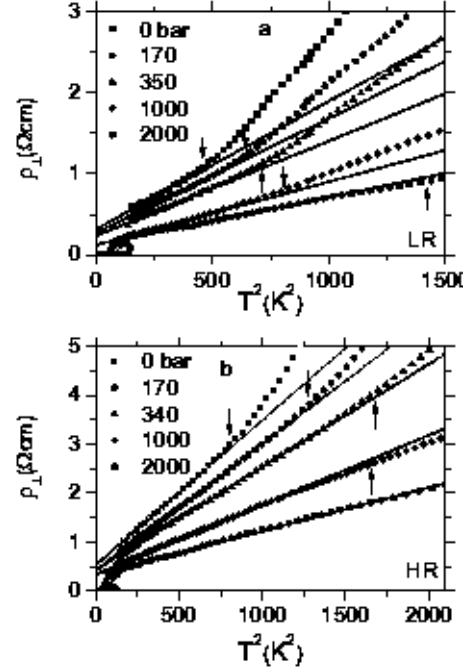

FIG. 5: Low-temperature out-of-plane resistivity data for the low (LR)(a)- and high (HR)(b)-resistance variant of κ-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Br under various pressures plotted as a function of T^2. Arrows mark the temperatures where the data deviate by more than 2% from the straight lines.

Figure 5 shows the low-temperature inter-layer resistivity data in a $\rho(T)$ vs. T^2 representation. In accordance with published results[21,23,24], the normal-state resistivity of the HR crystal (Fig. 5b) follows a $\rho(T) = \rho_0 + AT^2$ behavior over an extended temperature range. From Fig. 5b, we derive a coefficient $A^{HR} = (3 \pm 0.6)$ mΩcmK^{-2} and a residual resistivity $\rho_0^{HR} = (530 \pm 100)$ mΩcm. The error bars account for uncertainties implied in determining the geometrical factor. A T^2 dependence is also found for the LR crystal (Fig. 5a) although with markedly smaller values for the coefficient A and the residual resistivity of $A^{LR} = (1.6 \pm 0.4)$ mΩcmK^{-2} and $\rho_0^{LR} = (320 \pm 80)$ mΩcm, respectively. In addition, Fig 5 discloses significantly different validity ranges for the T^2 law for both variants. Using a 2% deviation of the straight lines in Fig. 5 as a measure for the upper boundary T_0 of the T^2 dependence, we find ambient-pressure values of $T_0^{LR} = (23 \pm 0.5)$ K and $T_0^{HR} = (30 \pm 0.5)$ K for the LR and HR crystal, respectively. As indicated by the arrows in Fig. 5, both variants reveal a strongly non-linear, and, for the HR crystal, even a non-monotonous, change of T_0 with pressure. We note that the analysis of the in-plane data at ambient pressure of the LR crystal in Fig. 1b reveals a T_0^{LR} value which is identical to that derived from the out-of-plane resistivity.
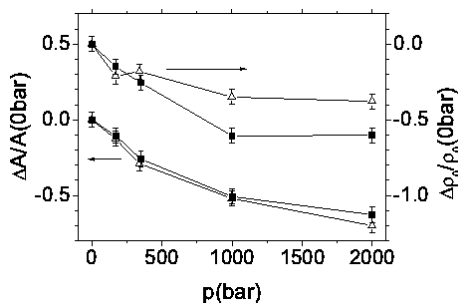
FIG. 6: Relative change of the coefficient A, $\Delta A = A(p)$ - A(0bar), and the residual resistivity ρ_0, $\Delta\rho_0 = \rho_0(p) - \rho_0(0bar)$, with pressure for the low (LR) (filled squares)- and high-resistance (HR) (open triangles) variant of single crystalline κ-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Br.

Figure 6 compiles the relative changes of the coefficient A, $\Delta A/A(p = 0) = A(0)^{-1}[A(p)$ - A(0)] (left scale) and the residual resistivity ρ_0, $\Delta\rho_0/\rho_0 = \rho_0^{-1}[\rho_0(p) - \rho_0(0)]$ (right scale) as a function of pressure. For the coefficient A, we find almost identical behavior for both crystals with a stronger reduction at small pressures and a weak pressure dependence at p $\geq$ 1 kbar. A similar tendency can be inferred also for the residual resistivity although here the pressure effect for the HR crystal is somewhat smaller and there is no significant pressure dependence for p $\geq$ 1 kbar.

IV. DISCUSSION

As described in the experimental section, the LR and HR variants of κ-(ET)$_2$Cu[N(CN)$_2$]Br are the results of somewhat different preparation routes which may cause variations in the purity of the materials, i.e. the concentration and the nature of incorporations[40], and the structural perfection. The latter refers to the degree and character of structural disorder. Although detailed comparative structural investigations of the LR and HR crystals which require highest-resolution techniques have not been performed yet, a general characterization of the crystals studied here is feasible on the basis of the present transport measurements.

According to the residual resistivities, derived from an extrapolation of the normal-state $\rho_\perp(T)$ to T = 0, of $\rho_0 = (320 \pm 80)$ mΩcm (LR) and (530 ± 100) mΩcm (HR), and the residual resistivity ratios RRR = $\rho_\perp(300K)/\rho_{0\perp}$ of 158 (LR) and 67 (HR), the LR crystal is of distinctly superior quality. The HR crystal studied here, however, appears to be representative for most of the κ-(ET)$_2$Cu[N(CN)$_2$]Br crystals studied so far which had been prepared according to the standard procedure. These crystals yield room-temperature resistivities $\rho_\perp(300K)$ and RRR values of 50-70 Ωcm and 50-65, respectively[19,21].

The possibility of internal strain, which might account for the suppression of the anomalous resistivity maximum

at intermediate temperatures for the LR crystal, can be safely discarded due to both the high T$_c$ value and the very narrow 10–90 % transition width of only 0.2 K. The latter is a factor of 2 smaller to that which is usually encountered for this salt[19,21] and which is found also for the present HR crystal. At the same time, both variants behave almost identical as for the glass-transition temperature T$_g$ = 77 K, although the signature at T$_g$ in the interlayer resistivity, i.e. the additional scattering contribution to $\rho_\perp$ for T < T$_g$, is stronger for the HR crystal. This might indicate a reduced fraction of frozen-in disordered ethylene groups in the LR compared to the HR sample.

The most obvious difference between the HR and LR crystals highlighted in figure 1 is the distinct reduction of the $\rho(T)$ maximum at intermediate temperatures. Yet a remnant of this feature, though much less strongly pronounced, is still present for the LR sample, where it gives rise to an unusual $\rho(T)$ anisotropy with a metallic-type resistivity in the out-of-plane component but a semiconducting-like behavior for the in-plane resistivity. We note, however, that the resistivity anisotropy $\rho_\perp/\rho_\parallel$ of about 100 at room temperature, derived from the present experiments (cf. Fig. 1) as compared to an anisotropy ratio in excess of 1000 reported by Buravov et al.[21], for an HR crystal employing an improved Montgomery method, indicates that the present $\rho_\parallel$ data determined by using a standard four-terminal measurement geometry still contain a significant interlayer component $\rho_\perp$.

A strongly reduced, though finite, scattering contribution around 90 K in the LR crystal is in line with the observation of a significant reduction of the still extraordinarily strong pressure response of the resistivity at intermediate temperatures compared to that of the HR crystal. In fact, sample-to-sample variations in this scattering contribution, though substantially reduced in size, may also be found for crystals prepared along the same alternative route that led to the present LR crystal. Here we mention the sample studied in reference [43] and two further crystals of this salt explored in the course of the present investigation. These samples revealed a clear correlation between the residual resistivity ratio and the size of the resistivity around 90 K: upon increasing the RRR ratio from 84 over 89 to 158, $\rho_\perp(90 K)$ continuously decreases. For the crystal studied in reference[43] yielding RRR = 193, $\rho_\perp(T)$ is almost identical to that found for the present LR crystal.

The above observation that the anomalous scattering contribution centered around 90 K differs strongly depending on the preparation conditions and becoming reduced in size upon increasing the sample quality, indicates that it is extrinsic in nature. Moreover, these results demonstrate that disorder or defects may induce drastic changes in the temperature-dependent part of the resistivity, i.e. the inelastic-scattering contributions. Such a behavior is highly unusual and at variance with what is known from ordinary metals, where the scatter-

ing due to disorder or impurities manifests itself in an increase of the residual resistivity only. This raises the fundamental question on how and to what extent disorder- or defect-induced potentials may affect the inelastic scattering of π electrons in the present molecular conductors.

Apart from these differences related to the anomalous resistivity contribution around 90 K, both variants behave identically as to the drastic change in their resistivity at T = T* $\simeq$ 40 K from a range characterized by a strongly T-dependent ρ(T) at T > T* into a low-temperature regime, where ρ(T) varies only weakly with temperature. It was found that the anomaly in dρ/dT coincides with the phase-transition-like feature observed in the coefficient of thermal expansion α(T). Such a direct correspondence of anomalies in transport and thermodynamic quantities is not expected for a crossover behavior between two different regimes, which usually involves a scaling factor to map the characteristic temperatures T$_\rho$ and T$_\alpha$. As an example, we mention the signatures of the Kondo effect in ρ(T) and α(T) in heavy fermion compounds, see, e.g.[42]. Rather the coincidence of distinct anomalies in dρ/dT and α(T) indicates that this feature marks a cooperative phenomenon.

Anomalous behavior around T* has been also identified in various thermal[7,20,21,25,26], magnetic[27,28,29,30], elastic[22], and optical properties[31]. Various explanations have been proposed as to the nature of the T* anomaly, including the formation of a pseudo-gap in the density of states[27,28,29,32], a crossover from a coherent Fermi liquid at low temperatures into a regime with incoherent excitations at high temperatures[4,14], a density-wave-type instability[20,26,33], as well as an incipient divergence of the electronic compressibility caused by the proximity to a Mott transition[5]. The present resistivity results, which for the HR crystal confirm published data[21], clearly demonstrate that the position of the T* anomaly is unaffected by the strength of the additional scattering contribution giving rise to the resistivity hump at intermediate temperatures, and thus marks an independent feature. In addition, the sharpness of the anomaly in dρ/dT and its direct mapping with the jump in the coefficient of thermal expansion makes it very unlikely that T* merely reflects a crossover between two different transport regimes—an assumption which underlies some of the above theoretical models. Rather it indicates that T* reflects a phase transition into a symmetry broken low-temperature state.

Turning now to the $\rho = \rho_0 + AT^2$ behavior for T $\leq T_0$ <T*, our study reveals a relative change with pressure of the coefficient A which is quite similar for the LR and HR crystals. This indicates that it is the same scattering mechanism which governs the low-temperature ρ(T) behavior for both systems. However, the size of A is substantially reduced for the LR crystal reflecting a weakening of this scattering contribution for the higher-quality LR crystal.

There has been a long-standing debate on the nature of the T^2 behavior in the resistivity of molecular conduc-

tors. In fact, a $\rho \propto T^2$ dependence over an extended temperature range is not a peculiarity of the κ-phase (ET)$_2$X salts alone. It has been observed also for various other materials such as the (TMTSF)$_2$PF$_6$ and the β-(ET)$_2$X salts, see, e.g.[34,35,36].

The explanations proposed for the T^2 behavior in these materials include electron-phonon[37] as well as electron-electron interactions of the strongly correlated π-electron system[4,14,34]. In fact, such a T^2 dependence at low temperatures is characteristic of metals in which the dominant scattering mechanism is provided by the electron-electron interactions. Since there the coefficient A $\propto$ $(m^*)^2$ $\propto$ $(T_F^*)^{-2}$, with m* the effective carrier mass and T$_F^*$ the effective Fermi temperature, the coefficient A scales with the square of the Sommerfeld coefficient $\gamma \propto m^* \propto (T_F^*)^{-1}$ of the electronic specific heat C$_{el}$ = γT. Such an A/γ^2 = const. behavior within a given material class has been verified for different systems including heavy-fermion compounds and transition metals[38,39].

The above scaling implies that upon variation of a control parameter x of the system, such as chemical composition or external pressure, the product A(x)$\cdot(T_F^*(x))^2$ should stay constant. By identifying the temperature T$_0$, i.e., the upper limit of the T^2 range in the resistivity, with the effective Fermi energy T$_F^*$, Limelette et al. have verified this invariance for pressurized κ-(ET)$_2$Cu[N(CN)$_2$]Cl[4].

The results of the present studies, however, render such an interpretation unlikely. Given that the T^2 dependence is of electronic origin, i.e. A $\propto (T_F^*)^{-2}$ and T$_0 \simeq T_F^*$, the A coefficient for the LR variant, which is reduced by a factor of about 1.9 compared to that of the HR crystal, would then indicate an effective Fermi temperature T$_F^*$ which is larger by a factor of $(1.9)^{1/2} \approx 1.4$. This is in contrast to the experimental observation yielding a T$_0^{LR}$ which is even reduced by a factor of about 1.3 compared to that for the HR crystal. Rather, our experimental finding that both T$_0$ and A are strongly sample dependent while the other characteristic temperatures associated with the electronic properties such as T* and T$_c$ are not, indicate that the nature of the T^2 dependence is different from coherent Fermi-liquid excitations[44].

V. SUMMARY

Resistivity measurements under hydrostatic pressure on a low-resistance variant of the organic superconductor κ-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Br have been performed and compared to the results on a standard high-resistance crystal. The lower residual resistivity ρ_0 and the higher residual resistivity ratio ρ(300K)/ρ_0 for the low-resistance crystal clearly indicate its superior quality. These measurements reveal that a significant part of the scattering contribution which gives rise to the anomalous resistivity maximum around 90 K in standard high-resistance materials is extrinsic in nature. Apart from this sample-dependent scattering contribution, however,

both variants behave identically as to the abrupt change in $\rho(T)$ at $T^* \simeq 40$ K. The coincidence of this temperature with the phase-transition anomaly in the coefficient of thermal expansion makes it unlikely that T^* marks a crossover between two different transport regimes but rather indicates a second-order phase transition. For temperatures $T \leq T_0 < T^*$ the data for both crystals were found to follow a $\rho(T) \propto AT^2$. Most importantly, however, our analysis reveals strikingly different coefficients A and ranges of validity measured by T_0 for both variants. In view of the fact that other characteristic temperatures associated with the π-electron system such as T_c and T^* are sample independent, this strong variation in A and T_0 indicates an origin for the T^2 dependence different from coherent Fermi-liquid excitations. The present results demonstrate that for these molecular materials, sample dependent, i.e. extrinsic factors such as disorder or defect concentration, does not only change the elastic scattering contribution measured by the residual resistivity. Rather, the defect potentials may also strongly affect the temperature-dependent part of the resistivity, i.e. the inelastic scattering, indicating that Matthiessen's rule is no longer applicable to these materials. Consequently, the charge transport for available sample materials might considerably be affected by such extrinsic contributions. Detailed structural investigations on high- and low-resistance material are in progress to hopefully identify the nature of the above scattering centers. This will help to control better the synthesis conditions and to eventually provide materials of sufficiently high quality which make it possible to access the intrinsic transport properties of these materials.

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